

IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

In re patent application of: STREBELLE, et al

Patent

Serial No.: 09/555,149

Examiner: Ildebrando, C.

Filed: May 26, 2000

Art Unit: 1754

For: EPOXIDATION CATALYST, ITS USE AND
EPOXIDATION METHOD IN THE PRESENCE
OF SAID CATALYST

Docket No.: P06745US0/BAS

DECLARATION UNDER 37 C.F.R. § 1.132

Commissioner for Patents
Washington, D.C.

S I R:

I, Michel Strebelle, declare that:

September 19th 2001
1. I am employed by Solvay ~~Interax~~ as a research chemist and I am a co-inventor of the present invention. I am also an inventor or co-inventor of numerous patents in this field, and I am well familiar with the subject matter of the present invention.

2. I have read and understood the Office Action dated September 26, 2001 and the prior art cited in that Office Action.

3. The present invention is directed to a process for producing an epoxidation catalyst based on titanium zeolite. The process includes blending a mixture comprising a titanium zeolite powder, water, at least one binder, at least one plasticizer, a pore-forming substance, and optionally other additives in order to form a paste. The paste is shaped by extrusion to form an extrudate. The extrudate is dried to remove at least some of the water and the extrudate is calcinated in order to remove at least some of the organic residues present. A granulation step is carried out between the extrusion and the drying step or after the calcinating step in order to obtain extruded granules.

4. None of the prior art references cited by the Examiner in the September 26, 2001 Office Action, namely Saxton et al, Muller et al, or Grosch et al, teaches or suggests the use of a pore-forming substance in the blend to be extruded.

5. Moreover, it was surprisingly and unexpectedly discovered that the addition of a pore-forming substance in accordance with the present invention results in a catalyst with a much higher activity while the catalyst still has a good resistance to attrition, as set forth in detail in the experiments described below.

Experiments

6. An experiment was performed using the present process. In the experiment, TS-1 powder was mixed with:

- a silicone binder in an appropriate amount to get the final SiO_2 content given in the table below
- 4g per 100 g TS-1 of a cellulose plasticizer
- 0, 10 or 20g per 100 g TS-1 of a pore-forming substance (melamine)
- 60g per 100 g TS-1 of water.

The mixture obtained was then extruded, calcined and granulated as in the example of the present application.

One part of the granules obtained was used in an attrition resistance test: 10g of the granules were inserted in a SpectroMill®-II Ball Pestle Impact Grinder from Chemplex®. After impact mixing during 10 min., the amount of fines (i.e., of particles smaller than 500 μm) was measured (by sieving and weighing).

Another part of the granules was used for the synthesis of EPI as in the example of the present application and the kinetic constant of reaction k (considering a first

reaction order in terms of H_2O_2 , i.e., reaction rate = k , $[\text{H}_2\text{O}_2]$ where $[\text{H}_2\text{O}_2]$ is the instant H_2O_2 concentration) was measured.

Results

7. The results of these experiments are summarized in the table below:

| Amount of SiO_2 after calcination (g/100g of TS-1) | Amount melamine of (*) (g/100g of TS-1) | Weight loss through attrition (%) | $k \cdot 10^3$ (min^{-1}) |
|---|---|-----------------------------------|--------------------------------------|
| 6 | 0 | 0.8 | 10 |
| 6 | 10 | 1.0 | 17 |
| 11 | 0 | 0.2 | 12 |
| 11 | 10 | 0.4 | 17 |
| 20 | 10 | 0.4 | 10 |
| 20 | 20 | 0.5 | 14 |

(*): before calcination, since all of it is removed during calcination; see present application, page 4, line 23.

Conclusion

8. The results shown in the table of paragraph 7 above demonstrate that a catalyst with an unexpectedly higher activity is obtained while the catalyst still has a good resistance to attrition.

I declare further that all statements made herein of my own knowledge are true and that all statements made on information and belief are believed to be true; and further that these statements were made with the knowledge that willful false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Date: December 13th
2001

Michel Strebelle
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